

# Nonequilibrium Kondo Effect in a Multi-level Quantum Dot near singlet-triplet transition

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The linear and nonlinear transport through a multi-level lateral quantum dot connected to two leads is investigated using a generalized finite- $U$  slave-boson mean field approach. For a two-level quantum dot, our calculation demonstrates a substantial conductance enhancement near the degeneracy point of the spin singlet and triplet states, a non-monotonic temperature-dependence of conductance and a sharp dip and nonzero bias maximum of the differential conductance. These agree well with recent experiment observations. This two-stage Kondo effect in an out-of-equilibrium situation is attributed to the interference between the two energy levels.

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Since the observation of the Kondo effect in semiconductor quantum dot (QD) with odd electron number [1], there have been a great deal of experimental and theoretical investigation into this many-body phenomenon. With easy control of major parameters of these artificial atoms in a wide range, QDs facilitate exploration of the Kondo physics at very different environments. As a consequence, new types of Kondo effects have been discovered in experiments. Very recently, a surprising Kondo-enhanced conductance has also been detected for an even number of electrons in both vertical [2] and lateral [3,4] QD configurations, which is markedly different from the conventional spin-1/2 Kondo effect.

A real QD contains more than one energy levels and usually two electrons incline to occupy the lower level, resulting in a singlet local spin state. Decreasing the spacing of the two energy levels by applying a magnetic field makes it possible for two electrons to occupy two different levels due to the exchange interaction  $J$ , forming the local spin-triplet state. Scaling theory [5,6] and numerical renormalization group analysis [7,8] revealed that this singlet-triplet transition gives rise to an anomalous enhancement of conductance at low temperature.

The physics in the lateral and vertical QD configurations are different. Recent observation on the lateral QD has disclosed a two-stage Kondo effect in the temperature-dependent conductance and differential conductance, and favored an interpretation in terms of a single conduction channel in two leads. [4] So far, there have been only a scaling analysis [6] and a numerical renormalization group calculation [8] on this problem in the low bias regime. Its clear physical picture and the nonlinear conductance at finite bias need further exploring. In this letter we investigate the singlet-triplet Kondo effect in a lateral QD having two energy levels in an out-of-equilibrium situation, by generalizing the finite- $U$  slave-boson mean-field (SBMF) approach, which was initially developed by Kotliar-Ruckenstein [9] and extended by us to study Kondo-type transport through QD [10] and coupled QDs [11]. Our results manifest not only a sub-

stantial enhancement of conductance near the local spin singlet-triplet transition, but also a two-stage Kondo effect in temperature-dependent conductance and differential conductance.

The Hamiltonian of the QD with two orbital energy levels ( $j = 1, 2$ ) connected to two leads ( $\eta = L/R$ ) can be written as  $H = H_L + H_R + H_D + H_T$ , in which  $H_\eta = \sum_{\eta,k,\sigma} \epsilon_{\eta k} c_{\eta k \sigma}^\dagger c_{\eta k \sigma}$  and  $H_T = \sum_{j,\eta,k,\sigma} (V_{j\eta} c_{\eta k \sigma}^\dagger c_{j\sigma} + \text{H.c.})$  describe the left and right leads and the tunneling between each lead and the QD (assuming a single conduction channel available per lead), respectively, and  $H_D$  represents the isolated QD ( $\alpha, \beta, \gamma, \sigma = \pm 1$ ):

$$H_D = \sum_{j,\sigma} \epsilon_j c_{j\sigma}^\dagger c_{j\sigma} + \sum_j U_j n_{j\uparrow} n_{j\downarrow} + U_{12} \sum_{\alpha\beta} n_{1\alpha} n_{2\beta} - J \sum_{\alpha\beta\gamma\sigma} c_{1\alpha}^\dagger c_{1\beta} c_{2\gamma}^\dagger c_{2\sigma} \boldsymbol{\sigma}_{\alpha\beta} \cdot \boldsymbol{\sigma}_{\gamma\sigma}, \quad (1)$$

where  $\epsilon_j$  in the first term is the single particle energy, the second and third terms denote the intra- and inter-level Coulomb interactions, and the last term gives a ferromagnetic exchange coupling  $J > 0$  due to Hund's rule. The energy spacing  $\delta = \epsilon_2 - \epsilon_1$  can be controlled by an external magnetic field. However, considering the small  $g$ -factor in, for example, GaAs, we neglect the Zeeman splitting in the QD, as done in previous theoretical treatment. [7,8] To keep the discussion simple we assume  $U_j = U_{12} = U$  and  $V_{j\eta} = V$  ( $j = 1, 2$  and  $\eta = L, R$ ).

Following the SBF scheme, [9] we introduce sixteen auxiliary Bose fields associated with sixteen single-particle eigenstates of the isolated multi-level QD Hamiltonian (1), as shown in Table I. For example, boson operator  $d_{1S_z}$  is with the spin-triplet state having spin ( $1, S_z$ ), and  $d_{01}$  is with the spin-singlet state  $|\uparrow\downarrow, 0\rangle$  (it is the lowest energy state in all three spin-singlet states). Note that the splitting of energies between this spin-singlet state and spin-triplet state  $\Delta = J - \delta$  can be tuned by a magnetic field. Therefore, for  $\delta = J$  the three triplet states  $d_{1S_z}$  and the singlet state  $d_{01}$  are degenerate. Necessarily, the completeness relation for these slave-boson operators,  $I = 1$  ( $I \equiv e^\dagger e + \sum_{j\sigma} p_{j\sigma}^\dagger p_{j\sigma} + \sum_{S_z \in \{1,0,\bar{1}\}} d_{1S_z}^\dagger d_{1S_z} +$

$\sum_{l=1}^3 d_{0l}^\dagger d_{0l} + \sum_{j\sigma} t_{j\sigma}^\dagger t_{j\sigma} + f^\dagger f$ ), and the condition for the correspondence between fermions and bosons,  $n_{j\sigma} \equiv c_{j\sigma}^\dagger c_{j\sigma} = Q_{j\sigma}$  ( $Q_{j\sigma} \equiv p_{j\sigma}^\dagger p_{j\sigma} + d_{1\sigma}^\dagger d_{1\sigma} + \frac{1}{2}d_{10}^\dagger d_{10} + d_{0j}^\dagger d_{0j} + \frac{1}{2}d_{03}^\dagger d_{03} + t_{j\sigma}^\dagger t_{j\sigma} + \sum_{\sigma'} t_{j\sigma'}^\dagger t_{j\sigma'} + f^\dagger f$  [ $\sigma, \sigma' = \pm 1$ ]), should be imposed to confine the enlarged Hilbert space. Moreover, in the combined fermion-boson representation, the QD fermion operators  $c_{j\sigma}^\dagger$  and  $c_{j\sigma}$  in the hopping term are expressed as  $z_{j\sigma}^\dagger c_{j\sigma}^\dagger$  and  $c_{j\sigma} z_{j\sigma}$ , respectively, where  $z_{j\sigma}$  consists of all the boson operator sets which are associated with the physical process that a  $\sigma$ -spin electron of the  $j$ th level is annihilated ( $\bar{j} \neq j, \bar{\sigma} \neq \sigma$ ):

$$z_{j\sigma} = Q_{j\sigma}^{-1/2} (1 - Q_{j\sigma})^{-1/2} \left[ e^\dagger p_{j\sigma} + p_{j\sigma}^\dagger d_{1\sigma} + \frac{1}{2} p_{j\bar{\sigma}}^\dagger (d_{10} + d_{03}) + p_{j\bar{\sigma}}^\dagger d_{0j} + d_{0j}^\dagger t_{j\sigma} + \frac{1}{2} (d_{10}^\dagger + d_{03}^\dagger) t_{j\bar{\sigma}} + d_{1\bar{\sigma}}^\dagger t_{j\bar{\sigma}} + t_{j\bar{\sigma}}^\dagger f \right]. \quad (2)$$

TABLE I. Sixteen Eigenstates, spin quantum numbers  $S$ ,  $S_z$  and energies  $E$  for the isolated QD with two levels 1 and 2, and the assigned slave-boson (SB) operators.

| Eigenstate  | $(S, S_z)$    | $E$                                   | SB                |
|---|---------------|---------------------------------------|-------------------|
| $ 0, 0\rangle$  | $(0, 0)$      | 0                                     | $e$               |
| $ \uparrow, 0\rangle$   | $(1/2, 1/2)$  | $\epsilon_1$                          | $p_{1\uparrow}$   |
| $ \downarrow, 0\rangle$   | $(1/2, -1/2)$ | $\epsilon_1$                          | $p_{1\downarrow}$ |
| $ 0, \uparrow\rangle$   | $(1/2, 1/2)$  | $\epsilon_2$                          | $p_{2\uparrow}$   |
| $ 0, \downarrow\rangle$   | $(1/2, -1/2)$ | $\epsilon_2$                          | $p_{2\downarrow}$ |
| $ \uparrow, \uparrow\rangle$  | $(1, 1)$      | $\epsilon_1 + \epsilon_2 + U - J$     | $d_{11}$          |
| $\frac{1}{\sqrt{2}}( \uparrow, \downarrow\rangle +  \downarrow, \uparrow\rangle)$ | $(1, 0)$      | $\epsilon_1 + \epsilon_2 + U - J$     | $d_{10}$          |
| $ \downarrow, \downarrow\rangle$  | $(1, -1)$     | $\epsilon_1 + \epsilon_2 + U - J$     | $d_{1\bar{1}}$    |
| $ \uparrow\downarrow, 0\rangle$   | $(0, 0)$      | $2\epsilon_1 + U$                     | $d_{01}$          |
| $ 0, \uparrow\downarrow\rangle$   | $(0, 0)$      | $2\epsilon_2 + U$                     | $d_{02}$          |
| $\frac{1}{\sqrt{2}}( \uparrow, \downarrow\rangle -  \downarrow, \uparrow\rangle)$ | $(0, 0)$      | $\epsilon_1 + \epsilon_2 + U + 3J$    | $d_{03}$          |
| $ \uparrow, \uparrow\downarrow\rangle$  | $(1/2, 1/2)$  | $2\epsilon_1 + \epsilon_2 + 3U - J$   | $t_{1\uparrow}$   |
| $ \downarrow, \uparrow\downarrow\rangle$  | $(1/2, -1/2)$ | $2\epsilon_1 + \epsilon_2 + 3U - J$   | $t_{1\downarrow}$ |
| $ \uparrow\downarrow, \uparrow\rangle$  | $(1/2, 1/2)$  | $\epsilon_1 + 2\epsilon_2 + 3U - J$   | $t_{2\uparrow}$   |
| $ \uparrow\downarrow, \downarrow\rangle$  | $(1/2, -1/2)$ | $\epsilon_1 + 2\epsilon_2 + 3U - J$   | $t_{2\downarrow}$ |
| $ \uparrow\downarrow, \uparrow\downarrow\rangle$                                  | $(0, 0)$      | $2\epsilon_1 + 2\epsilon_2 + 4U - 2J$ | $f$               |

Within the slave-boson scheme, the Hamiltonian of the system can be replaced by the following effective Hamiltonian in terms of auxiliary boson operators plus the constraints incorporated via the Lagrange multipliers  $\lambda$  and  $\lambda_{j\sigma}$ :

$$H_{\text{eff}} = \sum_{\eta, k, \sigma} \epsilon_{\eta k} c_{\eta k \sigma}^\dagger c_{\eta k \sigma} + \sum_{j, \sigma} \epsilon_j c_{j\sigma}^\dagger c_{j\sigma} + U \sum_j d_{0j}^\dagger d_{0j} + (U - J) \sum_{S_z \in \{1, 0, -1\}} d_{1S_z}^\dagger d_{1S_z} + (U + 3J) d_{03}^\dagger d_{03} + (3U - J) \sum_{j, \sigma} t_{j\sigma}^\dagger t_{j\sigma} + (4U - 2J) f^\dagger f + V \sum_{j, \eta, k, \sigma} (c_{\eta k \sigma}^\dagger c_{j\sigma} z_{j\sigma} + \text{H.c.}) + \lambda(I - 1) + \sum_{j, \sigma} \lambda_{j\sigma} (c_{j\sigma}^\dagger c_{j\sigma} - Q_{j\sigma}). \quad (3)$$

From the effective Hamiltonian (3) one can derive equations of motion of the slave-boson operators. Then we use the mean-field approximation in these equations and in the constraints, in which all the boson operators are replaced by their expectation values. The negligible Zeeman splitting helps to reduce the number of the independent expectation values, such that we have  $p_{j\sigma} = p_j$ ,  $d_{1S_z} = d_1$ ,  $t_{j\sigma} = t_j$ ,  $z_{j\sigma} = z_j$ , and  $\lambda_{j\sigma} = \lambda_j$ . With the help of the Langreth analytical continuation rules for the close time-path Green's function (GF), [12] these equations of motion can be closed in terms of the distribution GF  $G_j^<(\omega)$  of the QD, leading to the following self-consistent set of equations ( $j, j' = 1, 2$ ):

$$e^2 + 2 \sum_j p_j^2 + 3d_1^2 + \sum_{l=1}^3 d_{0l}^2 + 2 \sum_j t_j^2 + f^2 = 1, \quad (4)$$

$$\frac{1}{2\pi i} \int d\omega G_j^<(\omega) = p_j^2 + \frac{1}{2}(d_{03}^2 + 3d_1^2) + d_{0j}^2 + t_j^2 + 2t_j^2 + f^2, \quad (5)$$

$$\sum_j \frac{\partial \ln z_j}{\partial e} \mathcal{P}_j + \lambda e = 0, \quad (6)$$

$$\sum_{j'} \frac{\partial \ln z_{j'}}{\partial p_j} \mathcal{P}_{j'} + 2(\lambda - \lambda_j) p_j = 0, \quad (7)$$

$$\sum_j \frac{\partial \ln z_j}{\partial d_1} \mathcal{P}_j + 3(\lambda - \lambda_1 - \lambda_2 + U - J) d_1 = 0, \quad (8)$$

$$\sum_{j'} \frac{\partial \ln z_{j'}}{\partial d_{0j}} \mathcal{P}_{j'} + (\lambda - 2\lambda_j + U) d_{0j} = 0, \quad (9)$$

$$\sum_j \frac{\partial \ln z_j}{\partial d_{03}} \mathcal{P}_j + (\lambda - \lambda_1 - \lambda_2 + U + 3J) d_{03} = 0, \quad (10)$$

$$\sum_{j'} \frac{\partial \ln z_{j'}}{\partial t_j} \mathcal{P}_{j'} + 2(\lambda - \lambda_j - 2\lambda_{\bar{j}} + 3U - J) t_j = 0, \quad (11)$$

$$\sum_j \frac{\partial \ln z_j}{\partial f} \mathcal{P}_j + 2(\lambda - 2\lambda_1 - 2\lambda_2 + 4U - 2J) f = 0, \quad (12)$$

in which  $\mathcal{P}_j = \frac{1}{2\pi i} \int d\omega G_j^<(\omega) (\omega - \tilde{\epsilon}_j)$  and

$$G_j^<(\omega) = i \frac{\tilde{\Gamma}_j [f_L(\omega) + f_R(\omega)] (\omega - \tilde{\epsilon}_j)^2}{|\mathcal{D}(\omega)|^2}, \quad (13)$$

with

$$\mathcal{D}(\omega) = (\omega - \tilde{\epsilon}_1)(\omega - \tilde{\epsilon}_2) \pm i\tilde{\Gamma}_1(\omega - \tilde{\epsilon}_1) \pm i\tilde{\Gamma}_2(\omega - \tilde{\epsilon}_2), \quad (14)$$

$\tilde{\epsilon}_j \equiv \epsilon_j + \lambda_j$ , and  $\tilde{\Gamma}_j = \Gamma |z_j|^2$ ,  $\Gamma = \pi \sum_k |V|^2 \delta(\omega - \epsilon_{\eta k})$  being the coupling constant between the QD and the lead.  $f_{\eta}(\omega)$  is the Fermi distribution function of  $\eta$ th lead ( $\eta = L/R$ ). The current  $I$  through the two-level QD can be divided into three parts: the contributions from the ground level  $I_1$ , from the second level  $I_2$ , and the current  $I_i$  resulting from the interference between the two energy

levels. Straightforwardly, we have  $I = I_1 + I_2 + I_i = \frac{2e}{h} \int_{-\infty}^{+\infty} [T_1(\omega) + T_2(\omega) + T_i(\omega)][f_L(\omega) - f_R(\omega)]$ , in which

$$T_1(\omega) = \tilde{\Gamma}_1^2(\omega - \tilde{\epsilon}_2)^2 |\mathcal{D}(\omega)|^{-2}, \quad (15)$$

$$T_2(\omega) = \tilde{\Gamma}_2^2(\omega - \tilde{\epsilon}_1)^2 |\mathcal{D}(\omega)|^{-2}, \quad (16)$$

$$T_i(\omega) = 2\tilde{\Gamma}_1\tilde{\Gamma}_2(\omega - \tilde{\epsilon}_2)(\omega - \tilde{\epsilon}_1) |\mathcal{D}(\omega)|^{-2} \quad (17)$$

are the transmission probabilities.

In the linear limit the conductance can be written as

$$G = \frac{2e^2}{h} [T_1(0) + T_2(0) + T_i(0)] = \frac{2e^2}{h} \frac{(\tilde{\Gamma}_1\tilde{\epsilon}_2 + \tilde{\Gamma}_2\tilde{\epsilon}_1)^2}{|\mathcal{D}(0)|^2}. \quad (18)$$

In numerical analysis, we fix the parameters of the QD:  $U = 4$ ,  $J = 2$  with  $\epsilon_1 = -2.5$  or  $-3$ , to guarantee nearly two electrons dwelling in the QD (for  $-4 < \Delta < 1.2$ ,  $1.45 \lesssim N \lesssim 1.6$  at  $\epsilon_1 = -2.5$ ;  $1.5 \lesssim N \lesssim 1.7$  at  $\epsilon_1 = -3$ ), then focus our attention on the  $\Delta$ -dependent Kondo effect in transport. Here  $\Gamma$  is always taken as the energy unit and  $\epsilon_j$  is measured from the Fermi level of leads. Fig. 1 and the inset figure show the total linear conductance  $G$  and its three parts as functions of the energy splitting  $\Delta$  between the spin-singlet and -triplet states in the QD at zero temperature. There appears a peak in  $G$  near the singlet-triplet degeneracy point  $\Delta = 0$ . In the spin-singlet-state-dominated regime (singlet regime),  $\Delta < 0$ , the contribution of the second level,  $G_2$ , and that of the interference between the two levels,  $G_i$ , are almost zero. Only the first level is the active conduction channel. With increasing  $\Delta$  (decreasing  $\delta$ ), the second level starts to carry current, such that  $G$  rises up. Around the degeneracy point, the interference effect appears and its contribution  $G_i$  is negative. The rapid enhancement of the interference effect causes an abrupt decline of  $G$  in the spin-triplet-state-dominated regime (triplet regime)  $\Delta > 0$  and a sharp peak in  $G$  near the transition point. Note that the nonzero value of  $G$  in the singlet regime is determined by the position in the Coulomb blockade valley due to  $N < 2$  in our model QD parameters.

Fig. 2 illustrates the behavior of the conductance  $G$  as a function of temperature up to 0.3. It is believed that the SBMF approach is correct for describing the Kondo effect in strongly correlated systems at temperatures lower than the Kondo temperature  $T_K$ . For the systems under consideration, the Kondo temperatures  $T_K$  are estimated to be about 0.21 and 0.32 if only the first level is considered. [13] Therefore we can cautiously believe that the present calculation gives a rational description for the temperature dependence of the Kondo contribution to the conductance. In the cases  $\Delta = 0$  and 0.5, the conductance rises monotonously with decreasing temperature. In the cases  $\Delta = 1$  and 1.2, however, a ‘‘hump’’ behavior appears. After riseup with lowering temperature at first,  $G$  reaches a peak at a certain value of  $T$  before decreases, indicating a two-stage Kondo effect in this

triplet regime. These results are in qualitatively agreement with the scaling analysis of Pustilnik and Glazman [6], where a two-stage Kondo effect for an  $S \geq 1$  ground state (i.e., the triplet regime in this letter) was suggested. Moreover, we can see from Fig. 2 that more pronounced hump appears in deeper triplet regime (bigger  $\Delta$ ). It is also worth mentioning that, for moderate  $\Delta$ , for example,  $\Delta = 1$  at  $\epsilon_1 = -2.5$  [Fig. 2(a)] and  $\Delta = 0.5$  at  $\epsilon_1 = -3$  [Fig. 2(b)], the conductance  $G$  may slightly increase at very low temperatures, exhibiting a ‘‘shoulder’’ behavior, which satisfies very well with the recent measurements in Ref. [4] (inset of Fig. 2).

To simplify the calculation of nonlinear transport under a finite bias voltage between the two leads, we assume a symmetric voltage drop,  $\mu_L = -\mu_R = eV/2$ . The calculated zero-temperature current  $I$  as a function of bias voltage  $V$  in the case of  $U = 4$ ,  $J = 2$  and  $\epsilon_1 = -2.5$  are shown in Fig. 3(a) for  $\Delta \geq 0.5$ . With increasing the bias voltage in the range shown in the figure, 1) the current  $I$  increases monotonically for  $\Delta \lesssim 0.5$ ; 2)  $I$  approaches a peak value followed by a drop for  $\Delta > 0.5$ ; 3) the peak current appears at smaller bias for bigger  $\Delta$ . Correspondingly the voltage-dependent differential conductance  $dI/dV$  [Figs. 3(b)] shows a non-zero-bias-maximum at triplet regime  $\Delta > 0.5$ . While with decreasing  $\Delta$ , this sharp dip structure gradually transits to a conventional out-of-equilibrium Kondo behavior, i.e., zero-bias-maximum, at  $\Delta = 0.5$ . As shown in the insets in Figs. 2 and 3(b), where the separate contributions of the first and second levels as well as the interference term, to  $G$  and  $dI/dV$  are shown respectively, the hump behavior and the shoulder structure in  $G$ - $T$  curves, and the sharp dip in  $dI/dV$ - $V$  curves, can all be attributed to the strongly weakening of the interference effect due to increasing temperature or bias voltage.

In conclusion, we have generalized the finite- $U$  SBMF approach to the case of multi-level QD, and employed it to investigate the singlet-triplet Kondo phenomena in linear and nonlinear transport through a two levels QD in the  $N \simeq 2$  regime. Our investigation revealed a two-stage Kondo effect, a non-monotonic temperature-dependent conductance and a sharp dip structure in differential conductance, depending on the energy splitting  $\Delta$  between the spin-singlet and triplet states. These are in agreement with recent experimental observation. The predicted behavior of linear conductance is in consistent with existing theoretical analysis. An interpretation for this two-stage Kondo behavior in  $G$ - $T$  and  $dI/dV$ - $V$  has been provided in terms of the interference between the two levels.

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1.2. Inset in (b): Corresponding contributions of three parts  $dI_1/dV$ ,  $dI_2/dV$  and  $dI_i/dV$ .

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### Figure Captions

**Fig.1** Linear conductance at the absolute zero temperature as a function of the splitting between the spin-singlet and triplet states  $\Delta$  in QD of  $U = 4$  and  $J = 2$  with  $\epsilon_1 = -2.5$  and  $-3$ . Inset: Corresponding contributions of three parts  $G_1$ ,  $G_2$  and  $G_i$  for  $\epsilon_1 = -3$ .

**Fig.2** Temperature dependence of linear conductance with  $\Delta = 1.2, 1, 0.5$ , and  $0$  for (a)  $\epsilon_1 = -2.5$  and (b)  $\epsilon_1 = -3$ . Insets: Corresponding contributions of three parts  $G_1$ ,  $G_2$  and  $G_i$  for (a)  $\Delta = 1$  and for (b)  $\Delta = 1.2$ .

**Fig.3** (a) Zero-temperature  $I$ - $V$  characteristic curves for QDs with  $U = 4$ ,  $J = 2$ , and  $\epsilon_1 = -2.5$  at  $\Delta = 1.2 \sim 0.5$ . (b) Zero-temperature differential conductance  $dI/dV$  as a function of the external voltage at  $\Delta = 0.5, 1.0$ , and





